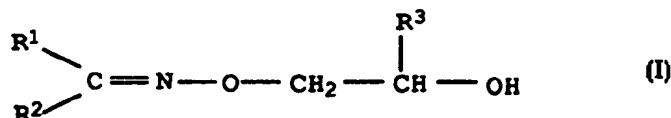




INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification 6 : C07C 249/12, 251/54	A1	(11) International Publication Number: WO 96/04238 (43) International Publication Date: 15 February 1996 (15.02.96)
(21) International Application Number: PCT/EP95/03001 (22) International Filing Date: 28 July 1995 (28.07.95) (30) Priority Data: P 44 27 289.8 ✓ 2 August 1994 (02.08.94) DE (71) Applicant (for all designated States except US): BASF AKTIENGESELLSCHAFT [DE/DE]; D-67056 Ludwigshafen (DE). (72) Inventors; and (75) Inventors/Applicants (for US only): HARREUS, Albrecht [DE/DE]; Teichgasse 13, D-67063 Ludwigshafen (DE). GÖTZ, Norbert [DE/DE]; Schöfferstrasse 25, D-67547 Worms (DE). RANG, Harald [DE/DE]; Ziegeleistrasse 76, D-67122 Altrip (DE). HARTMANN, Horst [DE/DE]; Lindenstrasse 45, D-67459 Böhl-Iggelheim (DE). MOHR, Jürgen [DE/DE]; Hochgewanne 48, D-67269 Grünstadt (DE). GEHRER, Eugen [DE/DE]; Londoner Ring 70, D-67069 Ludwigshafen (DE). LAUTH, Günter [DE/DE]; Stephensonstrasse 3, D-23562 Lübeck (DE). (74) Common Representative: BASF AKTIENGESELLSCHAFT; D-67056 Ludwigshafen (DE).		(81) Designated States: AU, BG, BR, BY, CA, CN, CZ, FI, HU, JP, KR, KZ, MX, NO, NZ, PL, RU, SG, SK, UA, US, UZ, European patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE). Published <i>With international search report.</i> <i>Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i>

(54) Title: PREPARATION OF O-(2-HYDROXYALKYL) OXIMES



(57) Abstract

Preparation of O-(2-hydroxyalkyl) oximes of formula (I), (R¹ and R² are alkyl or R¹ and R² and the common C atom are 5-membered to 7-membered cycloalkyl; R³ is alkyl) by causing a ketoxime II to react with an olefin oxide III in the presence of a tertiary amine or with a carbonate IV in the presence of a catalyst. The end products (I) are suitable for use as intermediates for crop protection agents.

INTERNATIONAL SEARCH REPORT

International Application No

PCT/EP 95/03001

A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 C07C249/12 C07C251/54

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 C07C

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	CHEMICAL ABSTRACTS, vol. 68, no. 11, 11 March 1968, Columbus, Ohio, US; abstract no. 49175, J. WOLF ET AL. page 4747 ; cited in the application see abstract & PL,A,53 525	1,5
X	--- CHEMICAL ABSTRACTS, vol. 73, no. 7, 17 August 1970, Columbus, Ohio, US; abstract no. 35231, J. WOLF ET AL. page 316 ; see abstract & PL,A,59 077 --- -/--	1,5

☒ Further documents are listed in the continuation of box C.☒ Patent family members are listed in annex.

* Special categories of cited documents :

- *A* document defining the general state of the art which is not considered to be of particular relevance
- *E* earlier document but published on or after the international filing date
- *L* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
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Date of the actual completion of the international search

22 November 1995

Date of mailing of the international search report

- 4. 12. 95

Name and mailing address of the ISA

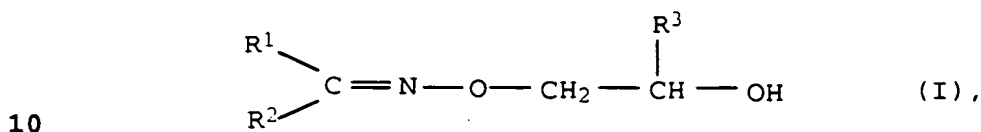
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Seufert, G

Preparation of O-(2-hydroxyalkyl)Oximes

The present invention relates to a process for the preparation of
5 O-(2-hydroxyalkyl)oximes of the general formula I



in which R¹ and R² each stand for an alkyl group having from 1 to
10 carbon atoms, or form, together with the carbon atom to which
they are attached, a 5-membered to 7-membered cycloalkyl radical,
15 and R³ denotes an alkyl group having from 1 to 10 carbon atoms.

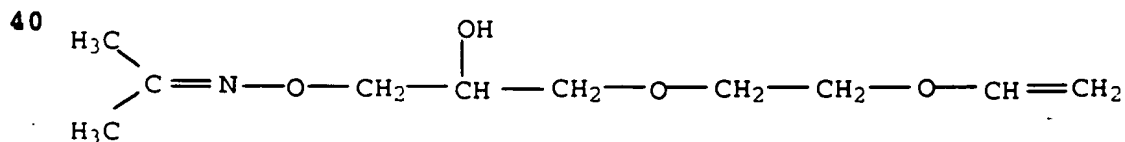
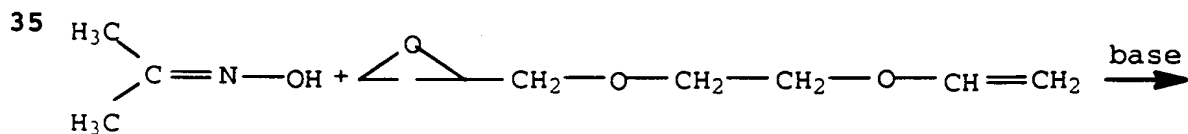
O-(2-hydroxyalkyl)oximes have great significance as intermediates
for plant protectants (cf eg, the prior German Application
DE-A 44 15 887).

20

J. Amer. Chem. Soc., 81, pp 4223 to 4225 (1959) and US-A
3,040,097 describe a process for the preparation of O-(2-hydroxy-
alkyl)-oximes in which the parent oximes are caused to react with
olefin oxides under basic conditions, in particular in the pres-
25 ence of alkali metal bases.

PL-A 53,525 (Chemical Abstracts, 68, 491 75d (1968)) reveals that
tertiary amines are suitable bases for a similar case.

30 Specially substituted O-hydroxyalkyloximes can be prepare by the
reaction of 25 glycol vinyl glycidyl ethers with oximes in the
presence of bases such as triethylamine or potassium hydroxide,
for example.



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(cf. Zh. Org. Khim., 23, 1426 to 1429 (1987)).

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Furthermore, it is well known that it is possible to cause reaction of acetone oxime with ethylene carbonate in toluene in the presence of potassium fluoride to produce O-(2-hydroxyethyl)acetone oxime (cf. Amer. Chem. Soc. Symposium Series 443 page 231(1991)).

Furthermore, a process for the preparation of bis[O-(2-hydroxyethyl)dimethyl-glyoxime starting from dimethylglyoxime and ethylene carbonate is described in J. Polymer Sci., 10, page 3408 (1972).

In accordance with the earlier application EP-A 655,437, aldoximes and ketoximes with unsubstituted or C₁-C₈-alkyl-substituted ethylene or propylene carbonate can be hydroxyalkylated in the presence of catalytic amounts of an N-alkylated, stable organic base or of pyridine substituted by a secondary amine.

It was the object of the invention to provide a more economical process for the preparation of O-(2-hydroxyalkyl)oximes I.

Accordingly, we have found a process for the preparation of O-(2-hydroxyalkyl)-oximes of the formula I, wherein a ketoxime of the formula II



is caused to react with

a) an olefin oxide of the formula III

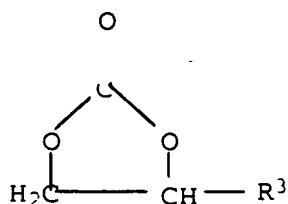


in the presence of a tertiary amine or

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b) a carbonate of the formula IV

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10 in the presence of a catalyst.

The reaction of II with a carbonate IV is preferred.

The following literature citations "Houben-Weyl" refer to Houben-
15 Weyl, Methoden der Organischen Chemie, 4th ed., Thieme Verlag, Stuttgart.

Preferred process products I are those which can be produced by
this process variant those are preferred in which R¹ and R² stand
20 for C₁-C₄ alkyl groups and primarily C₁-C₃ alkyl groups or form,
together with the carbon atom to which they are attached, a
cyclopentyl or cyclohexyl ring, and in particular those in which
R¹ and R² stand for methyl and/or ethyl or form, together with the
carbon atom to which they are attached, cyclohexyl. R³ preferably
25 stands for a C₁-C₄ alkyl group, particularly for ethyl and more
particularly for methyl. Most particularly preferred compounds I
are those in which R¹, R² and R³ stand for methyl.

The ketoximes II are generally known or are obtainable by known
30 methods, for example, by the reaction of the corresponding
ketones with hydroxylamine.

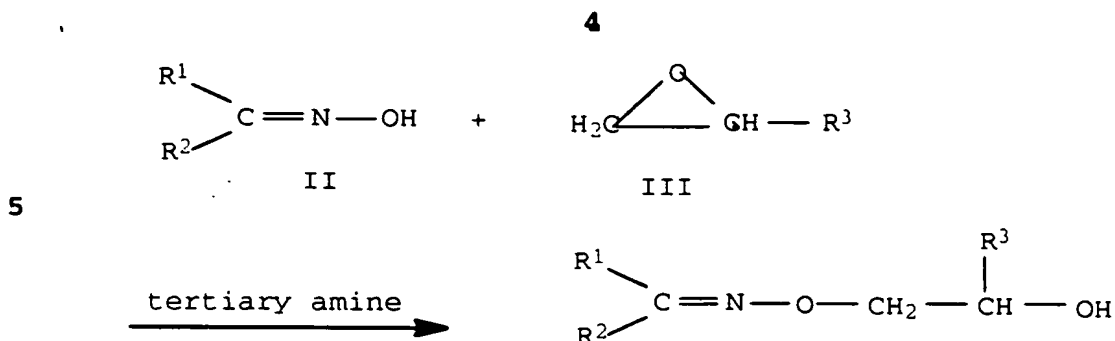
The olefin oxides III are also generally known or are obtainable
by known methods.

35

The carbonates IV are also generally known or can be produced by
known methods (cf. eg, EP-A 543,249).

(a) Reaction of ketoximes II with olefin oxides III in the pres-
40 ence of a tertiary amine:

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The molar ratio of olefin oxide III to ketoxime II is preferably from 1:1 to 2:1 and in particular from 1.1:1 to 1.3:1.

Suitable tertiary amines, either alone or in the form of a mixture, are primarily those having aliphatic and/or cycloaliphatic groups on the N atom such as triethylamine, tri-n-propylamine, tri-n-butylamine, tri-n-octylamine, tri-n-dodecylamine, N-methyldicyclohexylamine, and N,N-dimethylcyclohexylamine and also heterocyclic nitrogen bases having from 5 to 10 carbon atoms such as pyridine, 4-(N,N-dimethylamino)pyridine, 1,8-diazabicyclo[5.4.0]undec-7-ene, 1,4-diaza-bicyclo[2.2.2]octane, and 1-methylimidazole.

Preferably, with regard to the fact that the reaction mixture present at the end of the reaction is to be subjected to purification by distillation, a tertiary aliphatic amine is used whose boiling point is between 50° and 250°C and in particular between 80° and 180°C. Triethylamine and N,N-dimethylcyclohexylamine are particularly preferred.

30

The tertiary amine is usually employed in an amount of from 0.5 to 40 mol%, preferably from 0.9 to 10 mol% and more preferably from 1 to 5 mol%, based on the ketoxime II.

The reaction can be carried out without the use of solvent or in a suitable solvent or solvent mixture.

Suitable solvents are alkanols such as ethanol and tert-butanol, ethers such as 1,4-dioxane and tetrahydrofuran, ketones such as acetone, esters such as ethyl acetate, nitriles such as acetonitrile, N,N-dialkylamides such as dimethyl formamide and N,N-dimethylacetamide, N-alkylated lactams such as N-methyl-2-pyrrolidone, and N,N-dialkylated cyclic ureas such as N,N-dimethylethylene urea and N,N-dimethylpropylene urea.

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Also suitable as solvent is an excess of one of the aforementioned tertiary amines which is in the liquid state at least under the reaction conditions.

- 5 From a process engineering point of view, those of the aforementioned solvents are preferred, broadly speaking, which have a similar boiling point to that of the amines used and/or form azeotropes with them such that their removal, by distillation, from the crude reaction mixture takes place in a particularly
10 simple manner.

Particularly preferred are the N,N-dialkylamides and N-alkylated lactams, especially dimethylformamide and N-methyl-2-pyrrolidone.

- 15 There are usually employed from 50 to 500 mL and preferably from 50 to 200 mL of solvent, based on 1 mol of the ketoxime II.

The reaction is carried out at temperatures of from 40° to 130°C, and mostly from 70° to 110°C and pressures of from 0.5 to 40 bar
20 and mainly from standard pressure to 20 bar.

- Preferably the process is carried out by placing the ketoxime II and the tertiary amine, optionally in a solvent, in the reactor and metering in the olefin oxide III at the temperature of
25 reaction.

The reaction times are normally from 2 to 24 h but mostly from 4 to 16 h.

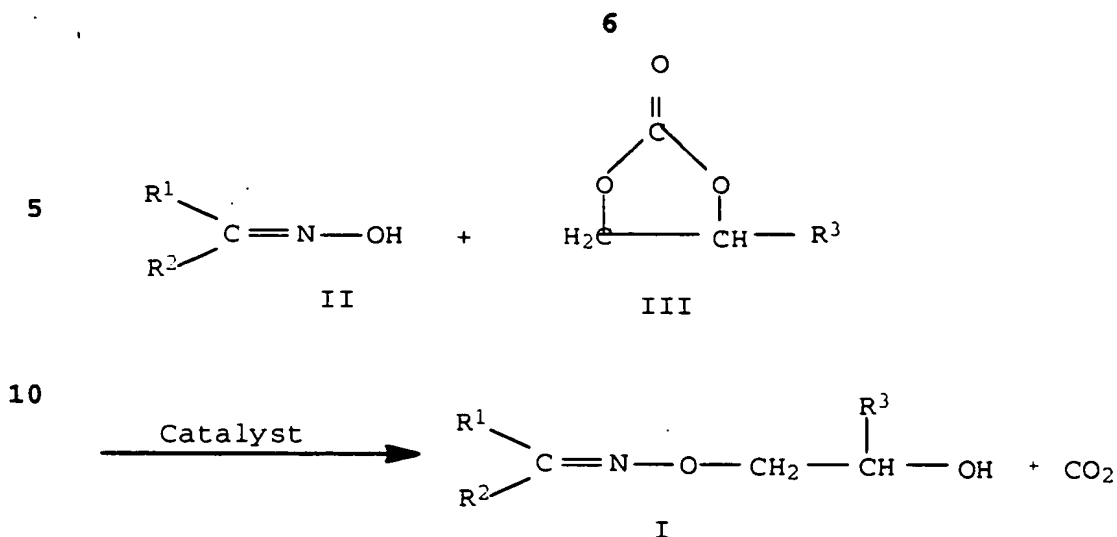
- 30 The isolation of the reaction products I takes place by methods known per se, preferably by distillation.

(b) Reaction of ketoximes II with carbonates IV in the presence of a catalyst:

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The molar ratio of carbonate IV to ketoxime II is usually from 1:1 to 10:1 and in particular from 4:1 to 7:1.

Suitable catalysts, used either alone or in the form of a mixture, are

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- 25
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- 35
- 40
- 45
- a) alkali metal salts, alkaline earth metal salts, and ammonium salts (NN_4^+), primarily the potassium salts and also the sodium salts' with inorganic or organic anions such as alkali metal halides, hydroxides, carbonates, hydrogen carbonates, alcoholates, and alkali metal salts of organic monocarboxylic acids. By way of example, there may be mentioned potassium fluoride, potassium iodide, potassium hydroxide, potassium carbonate, potassium hydrogen carbonate, potassium methyllate, potassium ethyllate, potassium tertbutyllate, and potassium acetate;
 - β) ammonium salts and preferably phosphonium salts having at least one organic group in the cation and containing inorganic or organic anions, primarily the halides, hydrogen carbonates, and acetates of such phosphonium salts in particular those containing three C_1 - C_4 alkyl and/or phenyl groups, such as tri-n-butylphosphonium acetate and triphenylphosphonium chloride;
 - ξ) phosphines having at least one but preferably three c-organic radicals, in particular those selected from the group comprising C_1 - C_{10} alkyl, C_6 - C_{15} -aryl, C_6 - C_{15} alkylaryl, and/or C_7 - C_{10} -arylalkyl, such as, primarily tri-n-butylphosphine and triphenylphosphine; or

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- δ) tertiary amines, preferably heterocyclics having from one to three N atoms as hetero atoms, such as, primarily, imidazoles, particularly N-(C₁-C₄ alkyl)imidazoles such as N-methylimidazole, and also triazoles, pyridines, and bicyclic azaheterocyclics such as 1,8-diazabicyclo[5.4.0]undec-7-ene and 1,4-diazabicyclo[2.2.2]octane.

Preferred catalysts are those which are mentioned under (α) and (δ). Of the (α) group, the potassium salts are preferably used, especially potassium fluoride or potassium hydrogen carbonate. Of the (δ) group, triethylamine is most particularly preferred.

The catalyst, primarily the inorganic alkali metal salts and the alkali metal salts of carboxylic acids, can be bonded to a support, if desired. Examples of suitable supports are aluminum oxide, silica gel, and kieselguhr. The amount of catalyst in these supported catalysts is usually from 10 to 50 wt%.

The catalysts are usually employed in an amount of from 5 to 50 mol%, preferably from 5 to 30 mol%, and more preferably from 5 to 10 mol%, based on the ketoxime II.

The reaction can be carried out in a single, preferably aprotic, organic solvent or in a solvent mixture. However, the process is preferably carried out without the use of a solvent, in which case an excess of the carbonate IV can, in particular, serve as reaction medium.

Suitable solvents are for example alkylbenzenes, such as, in particular, toluene and also the xylenes, furthermore dialkylketones such as methyl ethyl ketone, halobenzenes such as chlorobenzene, and ethers such as 1,4-dioxane.

The amount of solvent used is usually from 1 to 9 times and in particular from 2 to 7 times the weight of the ketoxime II. If the amount of solvent exceeds 9 times the weight of the ketoxime II, conversion and yield usually fall.

The reaction is preferably carried out at temperatures of from 80°C to 150°C and mostly from 100°C to 140°C and pressures of from 0.5 to 1 bar and especially under standard pressure.

The addition of a conventional phase transfer catalyst usually leads to an increase in reaction rate and conversion. However, for simple processing, it is preferred not to use a phase transfer catalyst.

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Examples of phase transfer catalysts are quaternary ammonium or phosphonium salts, preferably tetraalkylammonium, tetraalkylphosphonium, trialkylbenzylammonium, or trialkylbenzylphosphonium salts, especially triethylammonium, tributylbenzylammonium, and tetrabutylammonium chlorides, bromides, and hydrogen sulfates, and also tributylhexaphosphonium bromide.

The phase transfer catalyst is usually employed in an amount of from 0.5 to 2 mol% and preferably from 0.7 to 1 mol%, based on the ketoxime II.

The reaction can be carried out batchwise or, preferably, continuously.

The reaction times are normally from 8 to 24 h but mostly from 8 to 16 h.

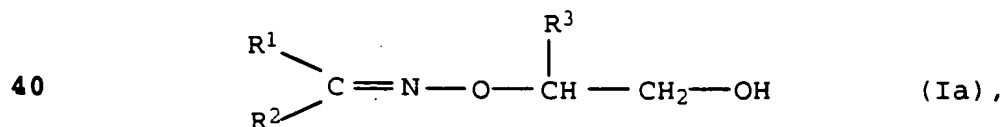
The isolation of the reaction product I takes place by methods known per se, preferably by distillation.

The distillation residues of the crude mixture contain substantially unconverted carbonate JV and the catalyst.

In a preferred embodiment of the process of the invention the process is carried out by replenishing the ketoxime II and the carbonate IV at a rate equal to the rate of consumption thereof, following the distillation of the product I from the reaction mixture, and the reaction is then repeated.

It has been found that in up to 20 successive reactions carried out in this manner in the same reactor, the yield of the compound I falls inappreciably and the ratio of the isomers I and Ia remains approximately the same.

The reaction frequently also produces, as by-product, the regioisomeric compound Ia



which is usually obtained in amounts of up to 10 %, based on I. The two isomeric compounds I and Ia are normally the first overheads occurring during distillation of the crude product. If desired, the isomer Ia can be substantially separated from the

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compound I by methods known per se, preferably by fractional distillation.

The end products I are important intermediates for crop protection agents, especially for herbicides of the cyclohexenone type (cf. DE-A 44 15 887).

Examples

10 Example 1

Preparation of O-(2-hydroxypropyl)propane-2-one oxime
(II + III according to (a))

- 15 2.92 kg (40 mol) of acetone oxime were dissolved in 5.2 L of dimethyl formamide and admixed with 0.2 kg (2 mol) of triethylamine. This mixture was heated to 70°C in an autoclave well purged with nitrogen gas. Over a period of 4 h, 2.56 kg (44 mol) of propylene oxide were metered in at this temperature, and on
- 20 completion of the addition, the reaction was allowed to continue for a further 3 h at 70°C. The autoclave was then depressurized, and unconverted propylene oxide was extensively removed at 200 mbar and 70°C, by distillation.
- 25 During purification, by distillation, of the crude mixture with the aid of a column having a length of 50 cm and a diameter of 5 cm and packed with 3 mm V₂A gauze rings, 2.84 kg of a mixture of O-(2-hydroxypropyl)propane-2-one oxime and O-(2-hydroxy-1-methylethyl)propan-2-one oxime were obtained at 78-81°C/30 mbar of. The
- 30 percentage isomer ratio I:Ia determined by gas chromatography was 96:4.

Example 2

- 35 Preparation of O-(2-hydroxybutyl)propan-2-one oxime
(II + III according to (a))

- Example 1 was repeated except that butylene-1 oxide was used instead of propylene oxide. During fractional distillation of the
- 40 crude product, a mixture of O-(2-hydroxybutyl)propan-2-one oxime and O-(2-hydroxy-1-ethylethyl)propan-2-one oxime was obtained at 80-82°C/20 mbar in a yield of 59 %. The isomer ratio of I:Ia was 96.7:3.3.

Example 3

Preparation of O-(2-hydroxypropyl)cyclohexanone oxime (II + III according to (α))

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Example 1 was repeated starting from cyclohexanone oxime instead of acetone oxime. During fractional distillation of the crude product a mixture of O-(2-hydroxypropyl)cyclohexanone oxime and O-(2-hydroxy-1-methylethyl)cyclohexanone oxime was obtained at 10 74°C/0.2 mbar in a yield of 54 %. The isomer ratio of I:Ia was 96:4.

Example 4

15 Preparation of O-(2-hydroxypropyl)propanone oxime (II + IV according to (β))

585 g (8 mol) of acetone oxime, 4080g (40 mol) of propylene carbonate, and 80 g (0.8 mol) of potassium hydrogen carbonate in 20 200g of toluene were caused to react for a period of 8 h at 130°C. There followed fractional distillation of the reaction mixture. The fraction of desired product consisting of O-(2-hydroxypropyl)propane-2-one oxime and O-(2-hydroxy-1-methylethyl)propan-2-one oxime distilled over at 70- 75°C/20-30 mbar in 25 a percentage ratio of I:Ia of 92:8.

Following cooling of the distillation residues, toluene, acetone oxime, and propylene carbonate were replenished to the aforementioned quantities and the reaction was then repeated. Following further 10 reactions of the type just described, an average 30 was taken over all 12 reactions. The yield of O-(2-hydroxyalkyl) oximes was found to be 90 % based on acetone oxime.

Example 5

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Preparation of O-(2-hydroxypropyl)propanone oxime (II + IV according to (β))

The process was carried out as in Example 4 but without the addition of toluene. Starting from 344 g (4.7 mol) of acetone oxime, 2420 g (23.5 mol) of propylene carbonate, and 47 g (0.47 mol) of potassium hydrogen carbonate there was obtained, in a total of 10 reaction cycles carried out, in each case, for a period of 8 h at 130°C, a mixture consisting of O-(2-hydroxypropyl)propane-2-one 45 oxime and O-(2-hydroxy-1-methylethyl)propane-2-one oxime in a percentage ratio of I:Ia of 92:8 and in a yield of 89%.

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Example 6

Preparation of O-(2-hydroxypropyl)propanone oxime
(II + IV according to (β))

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The process was carried out as in Example 4 but with the addition of 1086 g of toluene. Starting from 585 g (8 mol) of acetone oxime, 2450 g (24 mol) of propylene carbonate, and 80 g (0.8 mol) of potassium hydrogen carbonate there was obtained, in a total of 10 5 reaction cycles carried out, in each case over a period of 8 h at 125-130°C, a mixture consisting of O-(2-hydroxypropyl)propane-2-one oxime and O-(2-hydroxy-1-methylethyl)propan-2-one oxime in a percentage ratio of I:Ia of 92:8 and at a yield of 89 %.

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Example 7

Preparation of O-(2-hydroxypropyl)propane-2-one oxime
(II + IV according to (β))

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292.5 g (4.0 mol) of acetone oxime, 2040 g (20 mol) of propylene carbonate and 100 g (1.0 mol) of triethylamine were introduced into a stirred apparatus and stirred at an oil bath temperature of 130°C for 8 hours. After cooling, distillation was carried out 25 (bubble-cap tray column, 60 cm long, 30 mm in diameter). 80 g of triethylamine was recovered. The distillation residue, which consisted to the extent of 90% of unconsumed propylene carbonate, was able to be used for the next reaction with acetone oxime.

30 Yield (b.p. 9 mbar = 46°C): 85%.

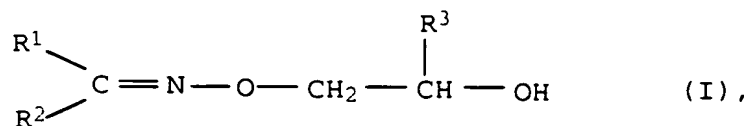
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We claim:

1. A process for the preparation of O-(2-hydroxyalkyl) oximes of
the general formula



in which R^1 and R^2 each stand for an alkyl group having from 1 to 10 carbon atoms or form, together with the carbon atom to which they are attached, a 5-membered to 7-membered cycloalkyl radical, and R^3 denotes an alkyl group having from 1 to 10 carbon atoms, wherein a ketoxime of the general formula II



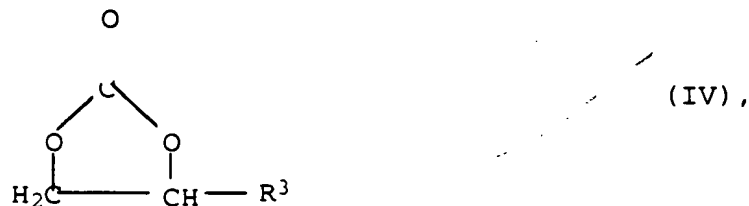
is caused to react with

- a) an olefin oxide of the general formula III



in the presence of a tertiary amine or

- b) a carbonate of the general formula IV



in the presence of a catalyst.

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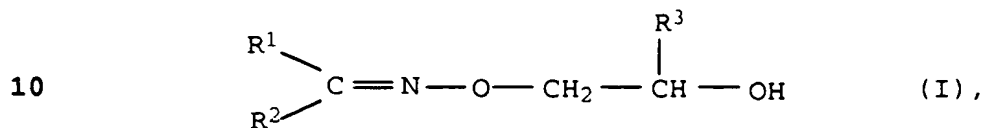
2. A process as defined in claim 1, wherein the ketoxime II used is acetone oxime, butanone oxime, or cyclohexanone oxime.
3. A process as defined in claim 1, wherein the ketoxime II used is acetone oxime.
4. A process as defined in any of claims 1 to 3, wherein the starting material is a compound III or IV in which R³ denotes methyl.
5. A process as defined in any of claims 1 to 4, wherein triethylamine is used as tertiary amine in process variant (a).
6. A process as defined in any of claims 1 to 4, wherein N,N-dimethylcyclohexylamine is used as tertiary amine in process variant (a).
7. A process as defined in any of claims 1 to 6, wherein potassium hydrogen carbonate is used as catalyst in process variant (b).
8. A process as defined in any of claims 1 to 6, wherein potassium hydrogen carbonate is used as catalyst in process variant (b).
9. A process as defined in any of claims 1 to 6, wherein a tertiary amine is used as catalyst in process variant (b).
10. A process as defined in claim 1, wherein II is reacted with IV without a solvent.

Preparation of O-(2-Hydroxyalkyl) Oximes

Abstract of the disclosure:

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Preparation of O-(2-hydroxyalkyl) oximes of the formula I



(R¹ and R² are alkyl or R¹ and R² and the common C atom are 5-membered to 7-membered cycloalkyl; R³ is alkyl) by causing a ketoxime II to react with an olefin oxide III in the presence of a tertiary amine or with a carbonate IV in the presence of a catalyst.

The end products I are suitable for use as intermediates for crop protection agents.

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INTERNATIONAL SEARCH REPORT

International Application No
PCT/EP 95/03001

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US,A,3 965 177 (L. E. KUNTSCHIK ET AL.) 22 June 1976 see column 3, line 31 - column 4, line 43 see column 5, line 6 - line 26 ---	1-6
X	R. KLAUS ET AL. 'ACS Symposium Series 443, 1991, Chapt. 18, p. 226-235', AMERICAN CHEMICAL SOCIETY, WASHINGTON cited in the application see page 231, scheme 9 ---	1
P,X	EP,A,0 655 437 (CIBA-GEIGY) 31 May 1995 cited in the application -----	1

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/EP 95/03001

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
PL-A-53525		NONE	
PL-A-59077		NONE	
US-A-3965177	22-06-76	NONE	
EP-A-0655437	31-05-95	CA-A- 2136497	26-05-95
		JP-A- 7188142	25-07-95
		US-A- 5434306	18-07-95

PCT

NOTIFICATION OF ELECTION

(PCT Rule 61.2)

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Date of mailing:

15 February 1996 (15.02.96)

International application No.:

PCT/EP95/03001

Applicant's or agent's file reference:

0050/45073

International filing date:

28 July 1995 (28.07.95)

Priority date:

02 August 1994 (02.08.94)

Applicant:

HARREUS, Albrecht et al

1. The designated Office is hereby notified of its election made:



in the demand filed with the International preliminary Examining Authority on:

15 December 1995 (15.12.95)



in a notice effecting later election filed with the International Bureau on:

2. The election ☒ was



was not

made before the expiration of 19 months from the priority date or, where Rule 32 applies, within the time limit under Rule 32.2(b).

The International Bureau of WIPO
34, chemin des Colombettes
1211 Geneva 20, Switzerland

Facsimile No.: (41-22) 740.14.35

Authorized officer:

J. Zahra

Telephone No.: (41-22) 730.91.11

PATENT COOPERATION TREATY

the
INTERNATIONAL PRELIMINARY EXAMINING AUTHORITY

PCT

ASF AG
Patentabteilung ZDX-C6
-67056 LUDWIGSHAFEN
ALLEMAGNE

Patentabteilung
- 6. NOV. 1996

NOTIFICATION OF TRANSMITTAL OF INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Rule 71.1)

Date of mailing
(day/month/year)

05. 11. 96

Applicant's or agent's file reference
0050/45073

IMPORTANT NOTIFICATION

International application No.

PCT/EP 95/03001

International filing date (day/month/year)

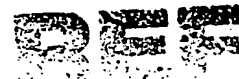
28/07/1995

Priority date (day/month/year)

02/08/1994

Applicant

BASF AKTIENGESELLSCHAFT et al.



The applicant is hereby notified that this International Preliminary Examining Authority transmits herewith the international preliminary examination report and its annexes, if any, established on the international application.

2. A copy of the report and its annexes, if any, is being transmitted to the International Bureau for communication to all the elected Offices.
3. Where required by any of the elected Offices, the International Bureau will prepare an English translation of the report (but not of any annexes) and will transmit such translation to those Offices.
4. REMINDER

The applicant must enter the national phase before each elected Office by performing certain acts (filing translations and paying national fees) within 30 months from the priority date (or later in some Offices)(Article 39(1))(see also the reminder sent by the International Bureau with Form PCT/IB/301).

Where a translation of the international application must be furnished to an elected Office, that translation must contain a translation of any annexes to the international preliminary examination report. It is the applicant's responsibility to prepare and furnish such translation directly to each elected Office concerned.

For further details on the applicable time limits and requirements of the elected Offices, see Volume II of the PCT Applicant's Guide.

Name and mailing address of the IPEA/



European Patent Office
D-80298 Munich
Tel. (+49-89) 2399-0, Tx: 523656 epmu d
Fax: (+49-89) 2399-4465

Authorized officer

Stefan Roche
S. Roche

Telephone No.

PCT

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference 0050/45073	FOR FURTHER ACTION See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. PCT/EP 95/ 03001	International filing date (day/month/year) 28/07/1995	Priority date (day/month/year) 02/08/1994
International Patent Classification (IPC) or national classification and IPC C07C249/12		
Applicant BASF AKTIENGESELLSCHAFT et al.		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.



2. This REPORT consists of a total of 8 sheets, including this cover sheet.

☐ This report is also accompanied by ANNEXES, i.e., sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consists of a total of _____ sheets.

3. This report contains indications and corresponding pages relating to the following items:

- I ☒ Basis of the report
- II ☐ Priority
- III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- IV ☒ Lack of unity of invention
- V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- VI ☒ Certain documents cited
- VII ☐ Certain defects in the international application
- VIII ☒ Certain observations on the international application

Date of submission of the demand 15/12/1995	Date of completion of this report 05. 11. 96
Name and mailing address of the IPEA/  European Patent Office D-80298 Munich Tel. (+49-89) 2399-0, Tx: 523656 epmu d Fax: (+49-89) 2399-4465	Authorized officer  C. Heibl Telephone No.

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

Intern. application No.

PCT/EP95/03001

I. Basis of the report

1. This report has been drawn up on the basis of (Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to the report since they do not contain amendments.):

☒ the international application as originally filed.

☐ the description, pages _____, as originally filed,
pages _____, filed with the demand,
pages _____, filed with the letter of _____,
pages _____, filed with the letter of _____.

☐ the claims, Nos. _____, as originally filed,
Nos. _____, as amended under Article 19,
Nos. _____, filed with the demand,
Nos. _____, filed with the letter of _____,
Nos. _____, filed with the letter of _____.

☐ the drawings, sheets/fig _____, as originally filed,
sheets/fig _____, filed with the demand,
sheets/fig _____, filed with the letter of _____,
sheets/fig _____, filed with the letter of _____.

2. The amendments have resulted in the cancellation of:

☐ the description, pages _____.
☐ the claims, Nos. _____.
☐ the drawings, sheets/fig _____.

3. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

4. Additional observations, if necessary:

IV. Lack of unity of invention

1. In response to the invitation to restrict or pay additional fees the applicant has:

- ☐ restricted the claims.
- ☐ paid additional fees.
- ☐ paid additional fees under protest.
- ☐ neither restricted nor paid additional fees.

2. ☐ This Authority found that the requirement of unity of invention is not complied with and chose, according to Rule 68.1, not to invite the applicant to restrict or pay additional fees.

3. This Authority considers that the requirement of unity of invention in accordance with Rules 13.1, 13.2 and 13.3 is

- ☐ complied with.
- ☒ not complied with for the following reasons:

The international preliminary examination report has been drawn up in respect of the entire international application but the international preliminary examining authority is of the opinion that the application does not comply with the requirements of unity of invention as set forth in the PCT regulations (Article 34(3), Rule 68(1) PCT).

The present process as defined in claim 1 comprises two different "process variants": (a) and (b).

The different reaction principles underlying both process variants are already basically known in the prior art, however.

For variant (a) see

- C.A. vol. 68, 49175 (1968)
- C.A. vol. 73, 35231 (1970)
- US-A 39 65 177

and the corresponding prior art further mentioned in

the present description on page 1;

For variant (b) see

- Am.Chem.Soc., ACS Symposium series 443, page 231
(1991)

and the corresponding prior art further mentioned on
page 2 of the description.

Having regard to this prior art, it is not apparent how
the process variants claimed in present claim 1 are
linked as to form a **single general inventive concept** as
required by Rule 13 PCT.

4. Consequently, the following parts of the international application were the subject of international preliminary examination in establishing this report:

☒ all parts.

☐ the parts relating to claims Nos. _____.

INTERNATIONAL PRELIMINARY EXAMINATION REPORTIntern. application No.
PCT/EP95/03001

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step and industrial applicability; citations and explanations supporting such statement

1. STATEMENT

Novelty (N)	Claims 1-10_____	YES
	Claims _____	NO
Inventive Step (IS)	Claims _____	YES
	Claims 1-10_____	NO
Industrial Applicability (IA)	Claims 1-10_____	YES
	Claims _____	NO

2. CITATIONS AND EXPLANATIONS

D1 C.A. vol. 68, 49175 (1968) & PL-A 53 525
D2 C-A. vol. 73, 35231 (1970) & PL-A 59 077
D3 Am.Chem.Soc. ACS Symposium Series 443, 1991,
Chapter 18, p.226-235
D4 EP-A 655 437

1. None of the prepublished documents mentioned in the Int. Search Report discloses the subject-matter of present claim 1 (and claims 2-10); the subject-matter is therefore novel (Art. 33(2) PCT).
(As for D4, which may become relevant at some stage, see section VI.)
 2. The present process as defined in present claim 1 in general terms comprises two different "process variants", viz. (a) and (b), which relate to basically known reaction methods:
as for variant (a) see D1, D2 and the corresponding further prior art teaching mentioned on page 1 of the
-

present description;
as for variant (B) see D3, page 231, scheme 9.

D3 teaches the addition of acetoneoxime to ethylene carbonate, catalysed by traces of potassium fluoride and tetramethylammonium chloride in refluxing toluene, which reaction is said to be "the simplest and most efficient possibility" to prepare the corresponding oximglycol. The desired product was obtained in a yield of 78,9%. The fact that when using propylene carbonate instead of ethylene carbonate under similar reaction conditions in accordance with the present application the product yields obtained are in the range of 85-90 % (see present examples 4-7) cannot be considered as a "surprising effect" supporting the presence of an inventive step. Moreover, as a rule less reactive reactants show higher selectivity; the selectivity however is proportional to the yield. It is further noted that it has been known to carry out the reaction described in D3 in the absence of a solvent (J.Polym.Sci. Part A-1, 1972,10,3405-19 mentioned in D4, page 1, lines 14-16).

In view of the prior art, it is not apparent how the process variants presently claimed are associated with an inventive step as required by Art. 33(3) PCT. No technical prejudice or obstacle can be seen which would have deterred the skilled person from attempting to produce the desired O-(2-hydroxyalkyl)-oximes of present general formula I by employing the known synthetic methods described in the cited prior art. Moreover, no "surprising effect" associated with the present process variants is recognisable from the application documents. The dependent claims 2-10 do not appear to contain any additional features which, in combination with the features of any claim to which they refer, involve an inventive step.

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

Intern. application No.

PCT/EP95/03001

VI. Certain documents cited

1. Certain published documents (Rule 70.10)

Application No. Patent No.	Publication date (day/month/year)	Filing date (day/month/year)	Priority date (valid claim) (day/month/year)
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EP-A 655 437	31.05.95	16.11.94	25.11.93
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Although EP-A 655 437 does not constitute prior art within the meaning of Rule 64.1(b), it appears to disclose all the features of process variant (b) of claim, cf. page 2, line 38 to page 5, line 5.

2. Non-written disclosures (Rule 70.9)

Kind of non-written disclosure	Date of non-written disclosure (day/month/year)	Date of written disclosure referring to non-written disclosure (day/month/year)
--------------------------------	--	---

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

Intern. application No.

PCT/EP95/03001

VIII. Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:

Claims 7 and 8 are identical.

REC'D 07 NOV 1996

WIPO PCT

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference 0050/45073	FOR FURTHER ACTION See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. PCT/EP 95/03001	International filing date (day/month/year) 28/07/1995	Priority date (day/month/year) 02/08/1994
International Patent Classification (IPC) or national classification and IPC C07C249/12		
Applicant BASF AKTIENGESELLSCHAFT et al.		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.


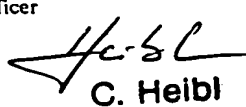
2. This REPORT consists of a total of 8 sheets, including this cover sheet.

☐ This report is also accompanied by ANNEXES, i.e., sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consists of a total of _____ sheets.

3. This report contains indications and corresponding pages relating to the following items:

- I ☒ Basis of the report
- II ☐ Priority
- III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- IV ☒ Lack of unity of invention
- V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- VI ☒ Certain documents cited
- VII ☐ Certain defects in the international application
- VIII ☒ Certain observations on the international application

Date of submission of the demand 15/12/1995	Date of completion of this report 05. 11. 96
Name and mailing address of the IPEA/  European Patent Office D-80298 Munich Tel. (+49-89) 2399-0, Tx: 523656 epmu d Fax: (+49-89) 2399-4465	Authorized officer  C. Heibl Telephone No.

I. Basis of the report

1. This report has been drawn up on the basis of (Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to the report since they do not contain amendments.):

☒ the international application as originally filed.

☐ the description, pages _____, as originally filed,
pages _____, filed with the demand,
pages _____, filed with the letter of _____,
pages _____, filed with the letter of _____.

☐ the claims, Nos. _____, as originally filed,
Nos. _____, as amended under Article 19,
Nos. _____, filed with the demand,
Nos. _____, filed with the letter of _____,
Nos. _____, filed with the letter of _____.

☐ the drawings, sheets/fig _____, as originally filed,
sheets/fig _____, filed with the demand,
sheets/fig _____, filed with the letter of _____,
sheets/fig _____, filed with the letter of _____.

2. The amendments have resulted in the cancellation of:

☐ the description, pages _____.
☐ the claims, Nos. _____.
☐ the drawings, sheets/fig _____.

3. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

4. Additional observations, if necessary:

IV. Lack of unity of invention

1. In response to the invitation to restrict or pay additional fees the applicant has:

- ☐ restricted the claims.
- ☐ paid additional fees.
- ☐ paid additional fees under protest.
- ☐ neither restricted nor paid additional fees.

2. ☐ This Authority found that the requirement of unity of invention is not complied with and chose, according to Rule 68.1, not to invite the applicant to restrict or pay additional fees.

3. This Authority considers that the requirement of unity of invention in accordance with Rules 13.1, 13.2 and 13.3 is

- ☐ complied with.
- ☒ not complied with for the following reasons:

The international preliminary examination report has been drawn up in respect of the entire international application but the international preliminary examining authority is of the opinion that the application does not comply with the requirements of unity of invention as set forth in the PCT regulations (Article 34(3), Rule 68(1) PCT).

The present process as defined in claim 1 comprises two different "process variants": (a) and (b).

The different reaction principles underlying both process variants are already basically known in the prior art, however.

For variant (a) see

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- C.A. vol. 73, 35231 (1970)
- US-A 39 65 177

and the corresponding prior art further mentioned in

the present description on page 1;

For variant (b) see

- Am.Chem.Soc., ACS Symposium series 443, page 231
(1991)

and the corresponding prior art further mentioned on
page 2 of the description.

Having regard to this prior art, it is not apparent how
the process variants claimed in present claim 1 are
linked as to form a **single general inventive concept** as
required by Rule 13 PCT.

4. Consequently, the following parts of the international application were the subject of international preliminary examination in establishing this report:

☒ all parts.

☐ the parts relating to claims Nos. _____.

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step and industrial applicability; citations and explanations supporting such statement

1. STATEMENT

Novelty (N)	Claims 1-10_____	YES
	Claims _____	NO
Inventive Step (IS)	Claims _____	YES
	Claims 1-10_____	NO
Industrial Applicability (IA)	Claims 1-10_____	YES
	Claims _____	NO

2. CITATIONS AND EXPLANATIONS

D1 C.A. vol. 68, 49175 (1968) & PL-A 53 525
D2 C-A. vol. 73, 35231 (1970) & PL-A 59 077
D3 Am.Chem.Soc. ACS Symposium Series 443, 1991,
Chapter 18, p.226-235
D4 EP-A 655 437

1. None of the prepublished documents mentioned in the Int. Search Report discloses the subject-matter of present claim 1 (and claims 2-10); the subject-matter is therefore novel (Art. 33(2) PCT).
(As for D4, which may become relevant at some stage, see section VI.)

2. The present process as defined in present claim 1 in general terms comprises two different "process variants", viz. (a) and (b), which relate to basically known reaction methods:
as for variant (a) see D1, D2 and the corresponding further prior art teaching mentioned on page 1 of the

present description;
as for variant (B) see D3, page 231, scheme 9.

D3 teaches the addition of acetoneoxime to ethylene carbonate, catalysed by traces of potassium fluoride and tetramethylammonium chloride in refluxing toluene, which reaction is said to be "the simplest and most efficient possibility" to prepare the corresponding oximglycol. The desired product was obtained in a yield of 78,9%. The fact that when using propylene carbonate instead of ethylene carbonate under similar reaction conditions in accordance with the present application the product yields obtained are in the range of 85-90 % (see present examples 4-7) cannot be considered as a "surprising effect" supporting the presence of an inventive step. Moreover, as a rule less reactive reactants show higher selectivity; the selectivity however is proportional to the yield. It is further noted that it has been known to carry out the reaction described in D3 in the absence of a solvent (J.Polym.Sci. Part A-1, 1972,10,3405-19 mentioned in D4, page 1, lines 14-16).

In view of the prior art, it is not apparent how the process variants presently claimed are associated with an inventive step as required by Art. 33(3) PCT. No technical prejudice or obstacle can be seen which would have deterred the skilled person from attempting to produce the desired O-(2-hydroxyalkyl)-oximes of present general formula I by employing the known synthetic methods described in the cited prior art. Moreover, no "surprising effect" associated with the present process variants is recognisable from the application documents. The dependent claims 2-10 do not appear to contain any additional features which, in combination with the features of any claim to which they refer, involve an inventive step.

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

Intern. application No.
PCT/EP95/03001

VI. Certain documents cited

1. Certain published documents (Rule 70.10)

Application No. Patent No.	Publication date (day/month/year)	Filing date (day/month/year)	Priority date (valid claim) (day/month/year)
EP-A 655 437	31.05.95	16.11.94	25.11.93

Although EP-A 655 437 does not constitute prior art within the meaning of Rule 64.1(b), it appears to disclose all the features of process variant (b) of claim, cf. page 2, line 38 to page 5, line 5.

2. Non-written disclosures (Rule 70.9)

Kind of non-written disclosure	Date of non-written disclosure (day/month/year)	Date of written disclosure referring to non-written disclosure (day/month/year)

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

Intern. application No.

PCT/EP95/03001

VIII. Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:

Claims 7 and 8 are identical.

INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference 0050/45073	FOR FURTHER ACTION see Notification of Transmittal of International Search Report (Form PCT/ISA/220) as well as, where applicable, item 5 below.	
International application No. PCT/EP 95/03001	International filing date (day/month/year) 28/07/95	(Earliest) Priority Date (day/month/year) 02/08/94
Applicant BASF AKTIENGESELLSCHAFT et al.		

This international search report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This international search report consists of a total of 3 sheets.

☒ It is also accompanied by a copy of each prior art document cited in this report.

1. ☐ Certain claims were found unsearchable (see Box I).
2. ☐ Unity of invention is lacking (see Box II).
3. ☐ The international application contains disclosure of a nucleotide and/or amino acid sequence listing and the international search was carried out on the basis of the sequence listing
 - ☐ filed with the international application.
 - ☐ furnished by the applicant separately from the international application,
 - ☐ but not accompanied by a statement to the effect that it did not include matter going beyond the disclosure in the international application as filed.
 - ☐ Transcribed by this Authority
4. With regard to the title, ☒ the text is approved as submitted by the applicant.
☐ the text has been established by this Authority to read as follows:
5. With regard to the abstract,
 - ☒ the text is approved as submitted by the applicant.
 - ☐ the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box III. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.
6. The figure of the drawings to be published with the abstract is:
Figure No. _____ ☐ as suggested by the applicant. ☐ None of the figures.
☐ because the applicant failed to suggest a figure.
☐ because this figure better characterizes the invention.

INTERNATIONAL SEARCH REPORT

International Application No
PCT/E5/03001A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 C07C249/12 C07C251/54

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 6 C07C

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	CHEMICAL ABSTRACTS, vol. 68, no. 11, 11 March 1968, Columbus, Ohio, US; abstract no. 49175, J. WOLF ET AL. page 4747 ; cited in the application see abstract & PL,A,53 525	1,5
X	--- CHEMICAL ABSTRACTS, vol. 73, no. 7, 17 August 1970, Columbus, Ohio, US; abstract no. 35231, J. WOLF ET AL. page 316 ; see abstract & PL,A,59 077 --- -/--	1,5

☒ Further documents are listed in the continuation of box C.☒ Patent family members are listed in annex.

* Special categories of cited documents :

- * "A" document defining the general state of the art which is not considered to be of particular relevance
- * "E" earlier document but published on or after the international filing date
- * "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- * "O" document referring to an oral disclosure, use, exhibition or other means
- * "P" document published prior to the international filing date but later than the priority date claimed

- * "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
- * "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
- * "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.
- * "&" document member of the same patent family

Date of the actual completion of the international search

22 November 1995

Date of mailing of the international search report

A. 5. 01

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2
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Tel. (+ 31-70) 340-2040, Tx. 31 651 epo nl,
Fax (+ 31-70) 340-3016

Authorized officer

Seufert, G

INTERNATIONAL SEARCH REPORT

International Application No

PCT/EP 95/03001

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US,A,3 965 177 (L. E. KUNTSCHIK ET AL.) 22 June 1976 see column 3, line 31 - column 4, line 43 see column 5, line 6 - line 26 ---	1-6
X	R. KLAUS ET AL. 'ACS Symposium Series 443, 1991, Chapt. 18, p. 226-235', AMERICAN CHEMICAL SOCIETY, WASHINGTON cited in the application see page 231, scheme 9 ---	1
P,X	EP,A,0 655 437 (CIBA-GEIGY) 31 May 1995 cited in the application -----	1

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/EP 95/03001

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
PL-A-53525		NONE	
PL-A-59077		NONE	
US-A-3965177	22-06-76	NONE	
EP-A-0655437	31-05-95	CA-A- 2136497	26-05-95
		JP-A- 7188142	25-07-95
		US-A- 5434306	18-07-95